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Life cycle assessment of flax shives derived second generation ethanol fueled automobiles in Spain

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ABSTRACT

Biofuel use seems to have certain environmental, energy and socioeconomic advantages versus fossil fuel consumption. The substitution of fossil fuels with biofuels can be a useful tool to fulfil the Spanish and European policy in relation to mitigation of greenhouse gas (GHG) emissions and increase the security in energy supply. The continuous increase in energy consumption, dependence on energy and high petroleum prices has motivated increasing support for renewable energy promotion. In Spain (the third ethanol producer in Europe in 2007), ethanol from lignocellulosic feedstocks could be one of the most valuable and interesting possibilities for renewable transportation fuels due to the limited competition with food production and high net reduction of GHG emissions. This study is focused on flax shives, obtained as an agricultural co-product from flax crops dedicated to fibre production for specialty paper pulp manufacture as lignocellulosic biomass to produce second generation ethanol involving the use of cellulosic technology. The life cycle assessment (LCA) methodology was used to evaluate the environmental impacts of the production and use in a flexi fuel vehicle (FFV) of ethanol blends (10 and 85% in volume of ethanol with gasoline) versus conventional gasoline, throughout their whole life cycle in order to highlight the main sources of these impacts. The system boundaries include cultivation, extraction, processing and final use of fuels. Mass and economic allocation were considered to determine the effect on the results of different allocation approaches.

The results of the study show that the allocation methods are essential for outcomes and decision-making. Using ethanol as transportation fuel could present better environmental performance than conventional gasoline in terms of global warming and fossil fuel consumption according to mass allocation. However, environmental credits could be achieved in terms of acidification, fossil fuel consumption and human toxicity according to economic allocation. Contributions to other impact categories such as eutrophication and photochemical oxidants formation were lower for conventional gasoline regardless of the allocation procedure selected. Agricultural activities related to feedstock production are notable contributors to the environmental performance. Thus, high yielding varieties, reduction of tillage activities and reduction in fertilization should help to reduce these impacts.

Contents

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1. Introduction

The progressive depletion of non-renewable fossil fuels and the increasing concern regarding climate change and atmospheric pollution, have conducted to an increased interest regarding renewable fuels use. Ethanol derived from biomass has the potential of being a sustainable transportation fuel as well as an alternative to gasoline [1,2]. In fact, it is used in the light duty vehicle fleet in a large number of countries [3]. Several authors have pointed out the environmental benefits of the use of biofuels (such as ethanol): reduction of primary fossil fuels use and greenhouse gas emissions [4–6] as well as energy requirement for ethanol production is lower than the energy content of ethanol produced [5,7,8].

Approximately 85% of the energy used in Spain is fossil fuel based. Spain is scarce in domestic energy sources and only coal is produced nationally in large amounts. Only 5.9% of total Spanish primary energy requirements as well as 16.6% of the total electricity production comes from renewable sources [9]. The promotion of renewable energies use is one of the principal vectors in the Spanish energy policy [10]. Spain was, in 2007, the third producer of ethanol in Europe behind France and Germany [11]. However, the use of ethanol only represents 0.4% of the total primary energy [9].

Traditionally, ethanol has been produced from starch and sugar crops such as cassava, rice, wheat, barley, corn grain or sugarcane [11,12]. However, a variety of biomass feedstocks can be used for ethanol production (second generation ethanol), e.g. mill wastes, municipal solid wastes and/or lignocellulosic biomass [13,14]. Lignocellulosic biomass includes materials such as herbaceous crops and agricultural and forest residues. Increase in starch prices, restrictions in the use of food crops for ethanol production, the abundance of lignocellulosic materials, the lack of competition with land and the shift from conventional ethanol to lignocellulosic ethanol, which does not require the redesign of completely new conversion plants, have made lignocellulosic biomass one of the most attractive feedstocks [15]. However, unlike starch, which contains homogenous and easily hydrolyzed polymers, these feedstocks are lignocellulosic, so they are not homogenous in nature [16].

Flax (*Linum usitatissimum*) is one of the most important fibre crops in Europe and in Northeast Spain [17,18]. This crop produces good quality non-wood fibres, which are used nowadays for speciality paper pulp, textile and automotive applications [19,20]. However, fibres constitute about 25–30% of the stem [20]. Flax shives are the woody and lignified inner tissues of the stem and are left over from processing stem into fibre. Both materials present a composition close to that of the wood species and are abundant renewable lignocellulosic materials with a potential conversion into ethanol and other high value products [20,21].

The most important product lines for shives are animal bedding (specifically horse bedding) and building (particleboards) [17,18,22]. In recent years, interest in the use of lignocellulosic content in shives as a source of fermentable sugars and phenolic compounds for the production of cellulosic ethanol and fine chemicals has grown due to their abundance and renewability [23].

Life cycle assessment (LCA) methodology has proven to be a valuable tool for analysing environmental considerations of product and service systems that need to be part of decision-making processes towards sustainability [24]. Several available LCA studies have identified the environmental performance of the production of ethanol from different feedstocks and the following use of the fuel in automobiles. Numerous studies have been published about production of ethanol from grains (corn and wheat) and its environmental performance in passenger cars [7,25]. The potential of corn stover as raw material to produce ethanol has been widely studied [2,6,16]. Sugar industry by-products e.g. cane molasses [26] and sugar crops such as sugar cane [27] and sugar beet [28] are examples of common feedstocks in tropical countries and environmental comparisons of using gasoline and ethanol based fuels have been made.

Cellulosic feedstocks such as switchgrass, salix and spruce were also analysed [2,16]. The analysis concluded that there is a potential reduction in greenhouse gas emissions when this kind of fuels is used. However, several differences have been found in the results due to the lack of a commercial cellulosic ethanol production line [2,29]. So far, although previous LCA studies have been conducted to assess the environmental impacts of cellulosic ethanol from cellulosic feedstocks, no studies were found on ethanol produced from flax shives.

This study focuses on flax shives obtained as an agricultural by-product from crops dedicated to fibre production for specialty paper pulp manufacture, as a second generation biofuel, involving the use of cellulosic technology.

A life cycle assessment (LCA) approach was used to evaluate the environmental impacts of a lignocellulosic biomass to ethanol process using flax shives as well as the use of ethanol as a transportation fuel (cradle to grave perspective) in a flexi fuel vehicle (FFV). In particular, the analysis compares the environmental performance of (i) ethanol in a 10% blend with gasoline (E10), (ii) ethanol in an 85% blend with gasoline (E85) and (iii) pure ethanol (E100) with unblended gasoline (CG).

A number of processes in the system under study yield more than one product such as the scutching process (long fibres, short fibres, shives and seeds). A sensitivity analysis was conducted in order to determine how the environmental results were affected by economic or mass allocation. Economic allocation according to the economic prices of flax products and co-products and mass allocation were taken into account.

2. Methodology

Environmental life cycle assessment (LCA) is defined as a methodology for the comprehensive assessment of the impact that a product has on the environment throughout its life cycle (from

extraction of raw materials through manufacturing, logistics and use to scrapping and recycling, if any), which is known as a "from cradle to grave" analysis [30]. LCA is an objective process to evaluate the environmental burdens associated to a product by identifying consumption of natural resources and emissions to environmental compartments, and to identify and implement opportunities to attain environmental improvements. The term product is broadly defined to include not only physical products or goods but also services and goods. According to the ISO 14040 [30], LCA is compiled of several interrelated components: (i) goal and scope definition; (ii) inventory analysis; (iii) impact assessment and (iv) interpretation of results for explanation of conclusions and recommendations, which is the scheme followed in this paper. LCA was used to compare the environmental performance of internal combustion engine automobiles fuelled with blends of gasoline and ethanol (E10, E85), pure ethanol (E100) and pure gasoline (CG), without local circumstances playing a role. Regarding the impact assessment stage, CML 1999 baseline impact assessment factors were chosen and modelling was performed using CMLCA (Chain Management by Life Cycle Assessment) [31].

2.1. Goal and scope definition

The goal of this study was to identify and compare the environmental performance of internal combustion engine automobiles fuelled with cellulosic ethanol and gasoline. Blend of 10% (by volume) ethanol with gasoline (E10) does not require engine modifications. However, the use of ethanol almost in its pure form

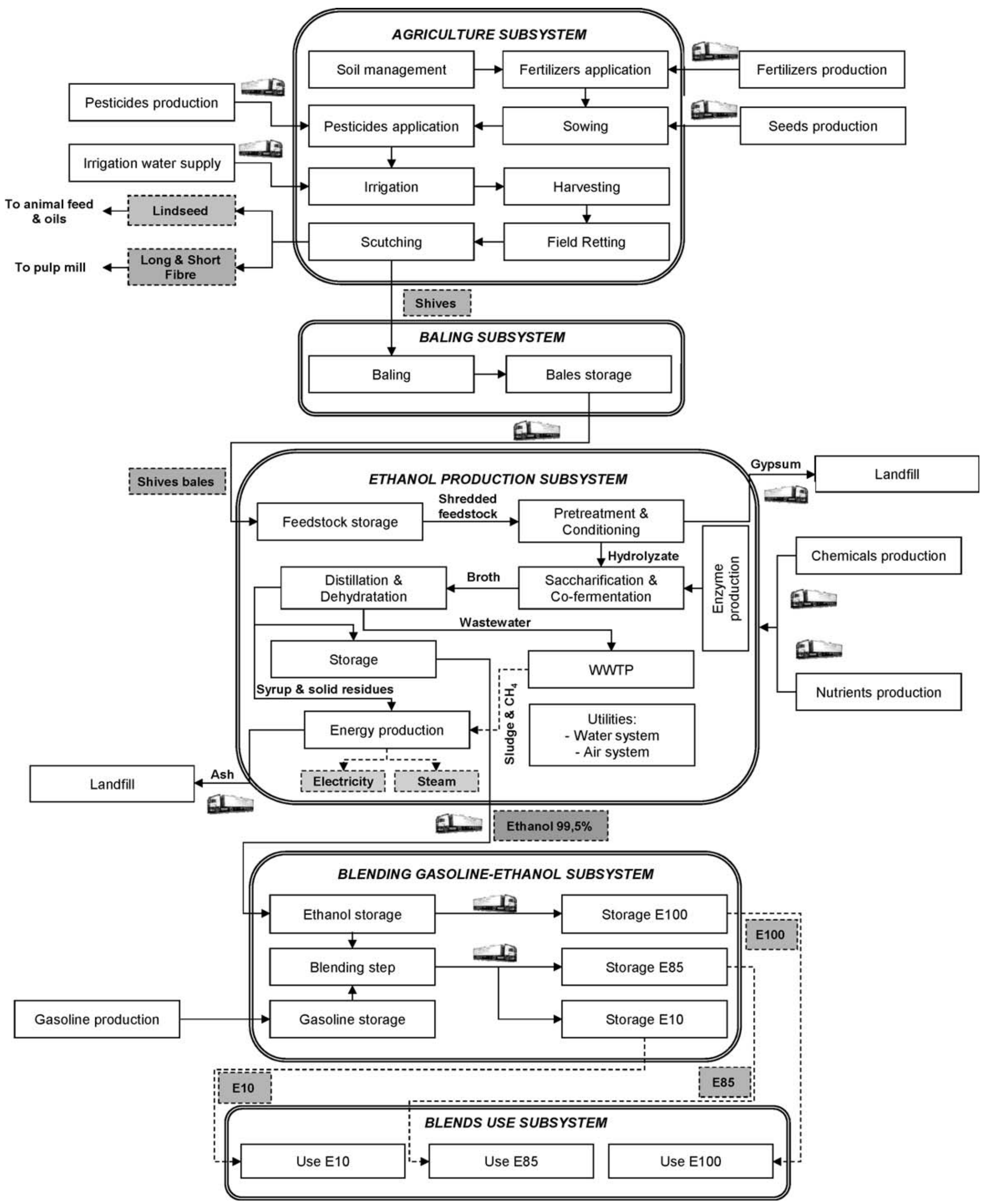

Fig. 1. System boundaries of flax shives based ethanol E10, E85 and E100 fuels life cycle.

(85–100%) (E85 and E100) requires modified engines. In this study, the use of flexi fuel vehicles (engines without modifications) was considered, which can use pure gasoline, pure ethanol or blends of both of them.

The scope of the study includes the life cycle of ethanol use from a cradle to grave perspective, including flax cultivation, shives treatment, ethanol conversion and transport to blending refinery, ethanol blending with gasoline and storage and, burning of fuel in FFV.

2.2. Functional unit

The functional unit provides the reference to which all inputs and outputs of the product system are related [30]. The functional unit chosen in this study to carry out the comparison between fuels was a distance of 1 km driven by a middle size FFV. In addition, 1 kg ethanol was considered as the functional unit when ethanol production was analysed in detail.

2.3. System definition and boundaries

The system under study was divided in five main subsystems: crop production (S1), bales formation (S2), ethanol refinery (S3), blending (S4) and fuel combustion in vehicles (S5), which are briefly described below. Fig. 1 shows a detailed description of the unit processes and subsystems considered within the system boundaries.

2.3.1. Agriculture subsystem (S1)

This subsystem includes not only all agricultural field operations for flax cultivation (ploughing, harrowing, fertilizing, pesticides application, sowing and harvesting), but also operations related to the post-preparation of the product (field retting, sun drying and scutching). The subsystem boundaries included the production of process materials such as fertilizers, pesticides and fuel for operating agricultural machinery production as well as their transportation to the farm gate. Spanish plantations of flax used for fibre production were taken into account.

2.3.2. Bales formation subsystem (S2)

Flax shives obtained from the scutching step are packed in bales and stored up for their final use. Shive bales are transported to ethanol refinery by 16 ton lorries. The average transport distance considered in this study was 180 km because this is the distance between plantations and the main pulp mill.

2.3.3. Ethanol refinery subsystem (S3)

Biological conversion of lignocellulosic feedstocks into ethanol is the process that receives the most attention and therefore, was the main process considered in this study [2]. The ethanol production materials and energy balances as well as ethanol yield were based on the ethanol conversion technology reported by the National Renewable Energy Laboratory [32] from corn stover, assuming that ethanol production efficiency is equal for other crops. In this case, feedstock composition was adapted to flax composition (Table 1).

In the ethanol conversion process, cellulose and hemicellulose are used for ethanol production. The lignin fraction of the biomass has a high heating value. It is used as fuel (as well as syrup, sludge and anaerobic biogas from the wastewater treatment plant, WWTP) in a lignin combustor in order to produce the energy requirements (heat and electricity) for the plant (self-sufficient). An overview of the ethanol process assumed is shown in Fig. 1. This subsystem was divided in nine processes: (i) feedstock handling and storage; (ii) pre-treatment and conditioning (where biomass is treated with dilute sulphuric acid and steam to liberate the hemicellulose sugars and other compounds; (iii) saccharification (or enzymatic hydrolysis) and co-fermentation; (iv) distillation and dehydration (including evaporation and solid–liquid separation) to purify and concentrate the ethanol up to 99.5%; (v) storage of ethanol; (vi) wastewater treatment plant (WWTP) where the bottoms of the distillation process and evaporator condensates are treated. Water treated is recycled to the refinery; (vii) energy production (electricity and process heat) from solids from distillation, syrup and biogas, (viii) enzyme production, where all enzymes required in the process are produced and finally (iv) ancillary utilities, which include the production of cooling, sterile and process water and compressed air. Gypsum from the solid–liquid separation and ashes from the energy production are sent to landfill.

Table 1
Assumed composition (dry basis) of feedstock delivered to the refinery gate.

Component	Weight fraction
Cellulose	0.477
Hemicellulose	0.170
Xylan	0.130
Arabinan	0.018
Other sugar polymers	0.022
Lignin	0.266
Acetate	0.030
Ash	0.010
Others	0.047
Total	1.00

2.3.4. Blending ethanol–gasoline subsystem (S4)

The production of gasoline as well as its transport to petrol stations was considered in this subsystem. In addition, gasoline and ethanol blending to produce the blends under study (E10 and E85) and their regional storage were also included within the subsystem boundaries. When pure gasoline and/or pure ethanol (E100) are used as fuels in a FFV, their delivery to a regional storage was also considered in this subsystem.

2.3.5. Blends use subsystem (S5)

Combustion of fuels described before in a representative FFV was evaluated and emissions were calculated according to the quantity of ethanol and gasoline necessary to drive 1 km. The environmental burdens associated with the vehicle production system and vehicle maintenance were not included in the analysis.

2.4. Inventory analysis

The most effort consuming step in the execution of LCA studies is the collection of inventory data in order to build the life cycle inventory (LCI). Moreover, high quality data is essential to make a reliable evaluation. The procedure for LCI of the system under study is summarized in Table 2, which is briefly described below. As observed, data for the study was collected from different sources and procedures.

Agricultural field activities (S1) were studied in detail and inventory data was obtained from expert advisors and personal communications with flax growers. The data was completed, when necessary, with bibliographic sources. Data for process materials consumed during the agricultural activities (fertilizers and pesticide) and energy carriers production was taken from the Ecoinvent database [33]. Transportation of inputs was included within the subsystem boundary, considering an average transport distance of 300 km by a 28-ton lorry [34]. However, neither transport of personnel to their workplaces nor the burdens from labour at farms were considered because it was not possible to

Table 2
Data sources for the life cycle inventory of lignocellulosic ethanol manufacture.

Subsystem	Data required	Data sources	Collecting method
S1	Fuel use Fertilizer use Pesticide use Labour use Consumable materials transport (mode, capacity and distance) Nutrient related emissions	Flax farmers, research reports [33–38]	Questionnaires, interviews, literature review
S2	Fuel use Weight of bales Shives bales transport (mode, capacity and distance)	Flax farmers, research reports [33,34], assumptions (see Table 4)	Literature review, questionnaires
S3	Production capacity Chemicals use Nutrients use Enzyme production Landfill operation Consumable materials transport (mode, capacity and distance) Energy requirements Industrial equipment use WWTP	Research reports [32,34,39–42], assumptions (see Table 4)	Literature review
S4	Gasoline production and transport (mode, capacity and distance) Ethanol transport (mode, capacity and distance) Ethanol, Gasoline and Blends storage	Research report [6,27,34], assumptions (see Table 4)	Literature review
S5	Fuel use Emission data of car driving	Research reports [43,44]	Literature review

collect representative data. The emissions of synthetic pesticides to air, surface water, groundwater and soil were estimated based on the methodology suggested by Hauschild [35]. Emissions related to fertilizer application were also calculated according to published reports [36–38].

Inventory data for the bales production subsystem (S2) were collected from published reports [33] and adapted to flax bale dimensions supplied by Spanish farmers. Average transport distance assumed was 180 km from bale storage to refinery gate. Emissions from the baling step, bale loading step into the lorry and transport were also considered in this subsystem. It can be forecasted that almost all emissions in this subsystem are derived from fossil fuel combustion in the engines.

Regarding the ethanol production subsystem (S3), the conversion of the dry biomass involves an enzymatic hydrolysis process followed by both fermentation and distillation processes. Design parameters and performance data for the ethanol production process was adapted to the selected feedstock composition and biomass capacity from the most recent ethanol conversion technology developed by the National Renewable Energy Laboratory of the USA Department of Energy [32] as described above. Inventory data was completed with emissions from the production of capital goods used in the refinery plant, taken from the EIPRO database [39]. Inventory data for the production of enzyme requirements was taken from [40]. Consumable material production was taken from Althaus et al. [41] and 50 km was assumed to be the average transport distance from wholesalers to refinery gate by means of 16 ton lorries. In addition, solid wastes generated in the ethanol production process such as ashes and/or gypsum were sent to landfill and 20 km was assumed as the delivery distance by means of 16 ton lorries. Inventory data for the landfill process was taken from Doka [42]. The refinery modelled in this study was considered to be energy self-sufficient. However, there is no electricity surplus to send to the national grid. The most relevant data for S3 is shown in Table 3.

Regarding the blending subsystem (S4), inventory data for the gasoline production and transport to the blending refinery was

Table 3
Global inventory for lignocellulosic ethanol production.

	Value
Inputs from the technosphere	
Materials	
Lignocellulosic feedstock (12.5% moisture)	43,680 kg
Vinyl acetate	12.5 kg
Sulphuric acid	1530 kg
Lime	1115 kg
Diammonium phosphate	64 kg
Corn steep liquor	498 kg
Enzyme	4003 kg
Nutrient feed	28 kg
Energy	
Electricity[a]	14,379 kWh
Steam[a]	176,586 MJ
Transport	
28 ton lorry	162.38 ton km
16 ton lorry	88.62 ton km
Inputs from the environment	
Materials	
Well water	85,842 kg
Outputs to the technosphere	
Materials	
Ethanol (99.5%)	11,325 kg
Wastes to treatment	
Gypsum (to landfill)	3340 kg
Ash (to landfill)	1090 kg
Outputs to the environment	
Emissions to air	
Vapour	102.79 ton
Acetic acid	46.70 ton
Carbon dioxide	46.32 ton
Ethanol	5.40 kg
Sulphuric acid	1 kg
Others	12.8 kg

[a] From energy production process from solids from distillation, syrup and biogas.

Table 4
Assumptions about delivery activities.

Materials	Transport mode	Capacity (ton)	Average distance (km)
Agrochemicals from wholesalers to farm gate	Diesel lorry	28	300
Shive bales from farm to refinery gate	Diesel lorry	16	180
Chemicals from wholesalers to refinery gate	Diesel lorry	16	50
Solid wastes from refinery to landfill gate	Diesel lorry	16	20
Ethanol from refinery to blending refinery gate	Diesel lorry	32	20
Blends to regional storage	Diesel lorry	32	34

taken from a published report [34]. Moreover, fuel distribution (pure gasoline, pure ethanol and, E10 and E85 blends) from the blending refinery to regional storage was assumed to be 34 km by 32 ton lorries. Emissions for the transport were also considered in this subsystem.

Information regarding the specific gravity and energy density properties of pure gasoline and ethanol necessary to estimate the efficiency of the engine running on E10, E85, E100 and gasoline were taken from Sheehan et al. [6].

Finally, emissions for the combustion of fuels under study in a FFV were taken into account in subsystem S5 and were taken from published reports [43,44]. Table 4 summarizes the assumptions regarding delivery activities related to ethanol production and use.

2.5. Allocation procedure

Allocation (partitioning of input or output flows of a unit process to the product under study) is one of the most critical issues in life cycle assessment and it was needed along this study as flax crops yield more than one product. Each allocation method (mass, economic, and energy) has advantages and disadvantages and choice of the allocation procedure depends on the limitations of the study.

In the flax crop under analysis, large amounts of shives are generated as a co-product during fibre separation. Lindseeds are also obtained and are commonly used for animal feed and oils. Economic allocation was initially applied as baseline according to the Handbook on Life Cycle Assessment [45] as fibre is the driving-force for this kind of cultivations and large differences in market prices are present [17,22,46]. The economic value of different agricultural products at the farm gate was used as basis for allocation. Large differences in the market prices for flax shives were identified, from 15 to 36 €/ton regardless of their final destination. Currently there is no available information concerning the price of the collected shives to ethanol production and therefore, it was assumed to be the same as the other shive uses. Feedstock cultivation has been identified as a notable contributor to the environmental impacts in the ethanol life cycle by several authors [3,29]. The dependence on the agricultural activities was analysed considering several scenarios in a sensitivity analysis. Three scenarios (EA1, EA2 and EA3) based on economic allocation were evaluated according to the differences in the market prices. Nevertheless, economic values can fluctuate since they depend on the EU subsidies for growers and their availability in markets. Thus, they could introduce uncertainty in the results. Therefore, mass allocation (scenario MA) was also assumed and considered in the sensitivity analysis in order to estimate the effects of the allocation factors on the environmental results, according to ISO 14044 [47].

The first scenario (EA1) is based on the lowest market value for the shives found in the literature [22]: 15 €/ton. The second one (EA2) is the base case and reflects the price paid nowadays for other lignocellulosic biomass for ethanol production e.g. corn stover [32]: 25 €/ton. The third one (EA3) assumes the highest market value for flax shives found in the literature [22]: 36 €/ton. Other authors have reported market value for flax shives around 20 €/ton (within our range) although large differences on the market prices for the other products have not been reported [22,46]. The last scenario (MA) is based on biomass production. A short description of the allocation factors considered in each scenario is shown in Table 5.

Regarding ethanol production, allocation was avoided because all the electricity produced from wastes in the refinery is consumed in the ethanol process. Therefore, there is no electricity surplus. In addition, solid residues generated in the refinery such as gypsum from distillation and ashes from boilers are sent to landfill and were considered as wastes. Thus, all the environmental burdens of the S3 were allocated to ethanol (the main product).

3. Environmental impact assessment

Among the phases defined by the impact assessment phase in the LCA methodology [30], only classification and characterization stages were considered. Normalization and evaluation were excluded since they are optional elements and according to the goal and scope defined here they would not provide extra useful information.

Characterization factors reported by the Centre of Environmental Science of Leiden University (CML baseline 1999 method) were considered [31], and the potential impact categories analysed are abiotic resources depletion (AD), global warming (GW), ozone

Table 5
Partitioning fraction for mass and economic allocation. Acronyms: MA = mass allocation; EAi = economic allocation i.

Product	Scenario							
	kg/ha	MA	€/kg	EA1	€/kg	EA2	€/kg	EA3
Long fibres	928	15.5%	2.181[a]	85.2%	2.181[a]	83.8%	2.181[a]	82.3%
Short fibres	569	9.5%	0.310[a]	7.4%	0.310[a]	7.3%	0.310[a]	7.2%
Shives	4048	67.5%	0.015[b]	2.6%	0.025[b]	4.2%	0.036[b]	5.9%
Linseed	453	7.6%	0.250[c]	4.8%	0.250[c]	4.7%	0.250[c]	4.6%
Total	6100	100%		100%		100%		100%

[a] Data from Ref. [22].
[b] Ref. [32].
[c] Ref. [46].

Table 6
LCA characterization results of ethanol production from flax shives per kg of ethanol. Acronyms: EAi = economic allocation i; MA = mass allocation.

Impact category	Unit	EA1	EA2	EA3	MA
AD	g Sb eq.	2.84	3.10	3.46	14.3
GW	kg CO_2 eq.	4.03	3.92	3.81	−0.367
OD	mg CFC-11 eq.	0.588	0.591	0.594	0.697
HT	g 1,4-DB eq.	11.1	131	15.4	961
FE	g 1,4-DB eq.	21.1	25.8	31.2	216
ME	kg 1,4-DB eq.	92.3	116	142	1070
TE	g 1,4-DB eq.	3.49	4.51	5.63	45.4
PO	g C_2H_4 eq.	1.76	1.81	1.87	3.80
A	g SO_2 eq.	4.28	4.59	4.91	16.7
E	g PO_4^{-3} eq.	0.721	0.984	1.26	11.4

Table 7
LCA characterization results for the impact categories under study based on 1 km distance driven by a FFV considering economic allocation (EA2) and mass allocation (MA).

Impact category	CG	EA2			MA		
		E10	E85	E100	E10	E85	E100
		% change relative to CG			% change relative to CG		
AD	1.69 g Sb eq.	−4.7	−66	−81	−1.2	−14	−16
GW	0.255 kg CO_2 eq.	+9.4	+95	+120	−2.7	−37	−46
OD	3.14×10^{-2} mg CFC-11 eq.	+6.4	+68	+87	+8.9	+94	+120
HT	14.5 g 1,4-DB eq.	−4.8	−4.1	−0.7	+37	+445	+565
FE	2.46 g 1,4-DB eq.	−4.5	+2.4	+5.7	+52	+607	+766
ME	12.5 kg 1,4-DB eq.	−5.6	−8.0	−7.2	+50	+594	+748
TE	0.281 g 1,4-DB eq.	+1.1	+46	+60	+107	+1188	+1501
PO	0.160 g C_2H_4 eq.	+2.5	+46	+56	+12	+144	+179
A	0.743 g SO_2 eq.	−3.6	−21	−22	+8.5	+107	+140
E	0.075 g PO_4^{-3} eq.	+2.8	+52	+73	+104	+1140	+1447

layer depletion (OD), human toxicity (HT), fresh water aquatic ecotoxicity (FE), marine aquatic ecotoxicity (ME), terrestrial ecotoxicity (TE), photochemical oxidants formation (PO), acidification (A) and eutrophication (E).

The environmental impacts addressed here reflect the differences between ethanol fuelled and gasoline fuelled vehicle operations and the influence of the allocation factors.

3.1. Environmental impact performance of ethanol manufacture

Table 6 summarizes the LCA characterization results (per kg of ethanol) for ethanol manufacture including subsystems S1, S2 and S3, and considering mass and economic allocation. Changes in the environmental impact results represent the effect of changing allocation factors for agricultural products. The allocation ratio between shives and the other co-products becomes higher when a change is performed from economic to mass allocation (from 0.026 in EA1 to 0.675 in MA). According to our results, a reduction in the environmental loads is only obtained in GW when the allocation ratio is really high; that is, when mass allocation was assumed as the case study. When the contributions to greenhouse gases (GHG) are analysed in detail, the emissions are due mostly to three global warming gases: CO_2 mainly from ethanol production, N_2O from the cultivation phase (direct and indirect emissions from nitrogen based fertilizer application) and followed by far by CH_4 from fossil fuel extraction (Fig. 2). In addition, it is important to remark the positive effect of the carbon sequestered during crop growth (~9.9 ton CO_2/ha), which contributes to offset the GHG emissions. This effect is more outstanding in the mass allocation (highest allocation factor) since more CO_2 taken up during the crop growing is allocated to flax shives. Although more N_2O and CH_4 emissions are also allocated to shives, the net value in CO_2 equivalents is reduced. The most relevant processes which contribute to the GW are shown in Fig. 3. Activities related to the ethanol conversion plant, such as distillation and electricity production, are the main hot spots in this impact category. In addition, when mass allocation is assumed, there is a remarkable contribution from fossil fuel extraction due to a higher amount of diesel from agricultural machineries being allocated to the flax shives.

Concerning the remaining categories analysed (AD, OD, HT, FE, ME, TE, PO, A and E), the contributions increase when allocation ratio is increased. These results are mostly due to agricultural activities. Manufacture of fertilizers and agricultural machinery production are the main contributors to almost all of them, excluding AD and OD. In both these categories, the extraction of fossil fuels extraction, such as natural gas, coal and oil, are the main contributors. The largest value obtained in MA is related to more diesel consumed in agricultural activities. It is allocated to ethanol.

Feedstock culture contributes significantly to A, E and PO due to SO_2 emissions from fertilizers manufacture (in particular, P based fertilizers), N_2O from N based fertilizer application and, NO_x, CO and NMVOC emissions from diesel combustion in agricultural machineries as well as fertilizer production.

3.2. Environmental impact performance of E10, E85 and E100

3.2.1. Comparison between ethanol blends

Two allocation approaches were selected and analysed in detail in order to identify the effects on the environmental results of substituting any of the three alternatives (E10, E85 and E100) for CG: economic allocation 2 (EA2) since it is the base case and presents the current situation in terms of market prices for the

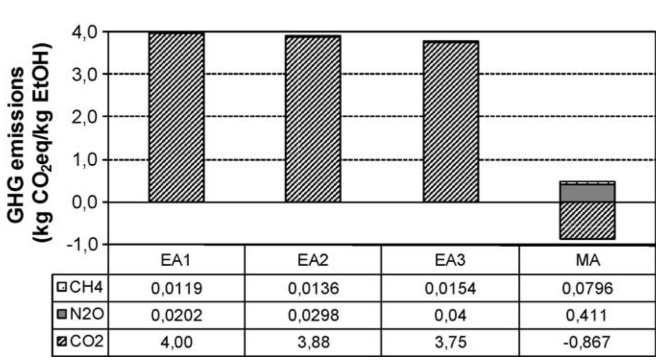

Fig. 2. Comparative GHG emissions for ethanol production according to the allocation procedures. Acronyms: EA1 = economic allocation 1; EA = economic allocation 2; EA3 = economic allocation 3; MA = mass allocation.

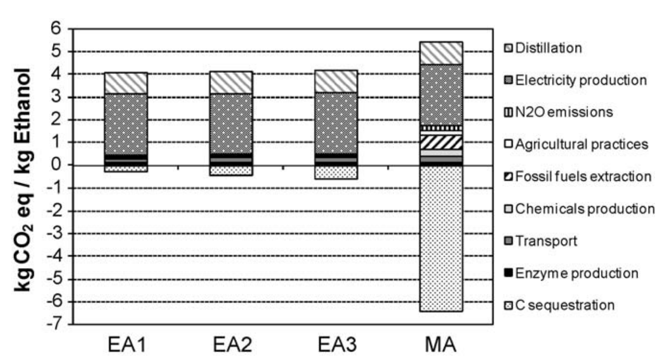

Fig. 3. Comparison (between allocation factors) of life cycle CO_2 equivalent emissions for ethanol production and main processes involved. Acronyms: EA1 = economic allocation 1; EA = economic allocation 2; EA3 = economic allocation 3; MA = mass allocation.

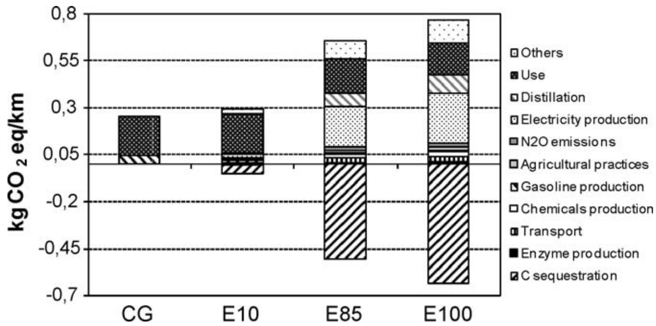

Fig. 4. Contributions of the main processes involved to GW, based on mass allocation scenario (MA).

lignocellulosic biomass for ethanol manufacture and mass allocation (MA) because it presents the highest allocation factor (67.5%).

Table 7 presents the life cycle assessment characterization results for E10, E85, E100 and CG. Changes represent impacts of substituting the fuel alternative for CG. Negative changes imply a reduction in the environmental loads compared to gasoline, whilst positive changes denote an increase.

For the EA2 scenario, the results show that the environmental performance of the ethanol based fuels application is better than the use of CG in terms of AD, HT, ME and A. These favourable effects are mainly caused by both the negligible contribution from agricultural activities due to the low allocation factor and the low ratio of gasoline in the blends (mainly in E85 and E100). In the remaining impact categories (GW, OD, FE, TE, PO and E), shifting from CG to ethanol blends does not present environmental benefits. On the one hand, the low allocation factor implies the reduction in the CO_2 taken up during the biomass growth assigned to the feedstock, which leads to an increase in the GW. On the other hand, these unfavourable environmental effects are caused by the environmental burdens related to upstream activities, especially from the ethanol conversion stage, which increase when the ethanol ratio in the blend is.

Regarding the MA scenario (Table 7), the tendency is the same as EA2 in almost all impact categories excluding GW, ME and A, although the net contributions increase in comparison with EA2 due to the higher contribution from feedstock cultivation. The use of ethanol as liquid fuel in a FFV only results in environmental credits for GW and AD. Agricultural activities are responsible for the reduction of contributions of AD in comparison to EA2, mainly due to the fact that more diesel requirements from agricultural activities are allocated to the shives. Nevertheless, the reduction of fossil fuel extractions is increased when ethanol ratio in the blend is increased. This is due to the avoidance of using gasoline as a transport fuel.

The tendency of the results is completely opposite in terms of GW, ME and A. The results in terms of GW are derived from the absence or less consumption of gasoline in E100, E85 and E10 and consequently, the increase in the CO_2 taken up from flax growth. The evaluation of GHG emissions included the emissions due to fossil and non-fossil fuels use (there is no demand for fossil fuels in the ethanol production subsystem since process energy source

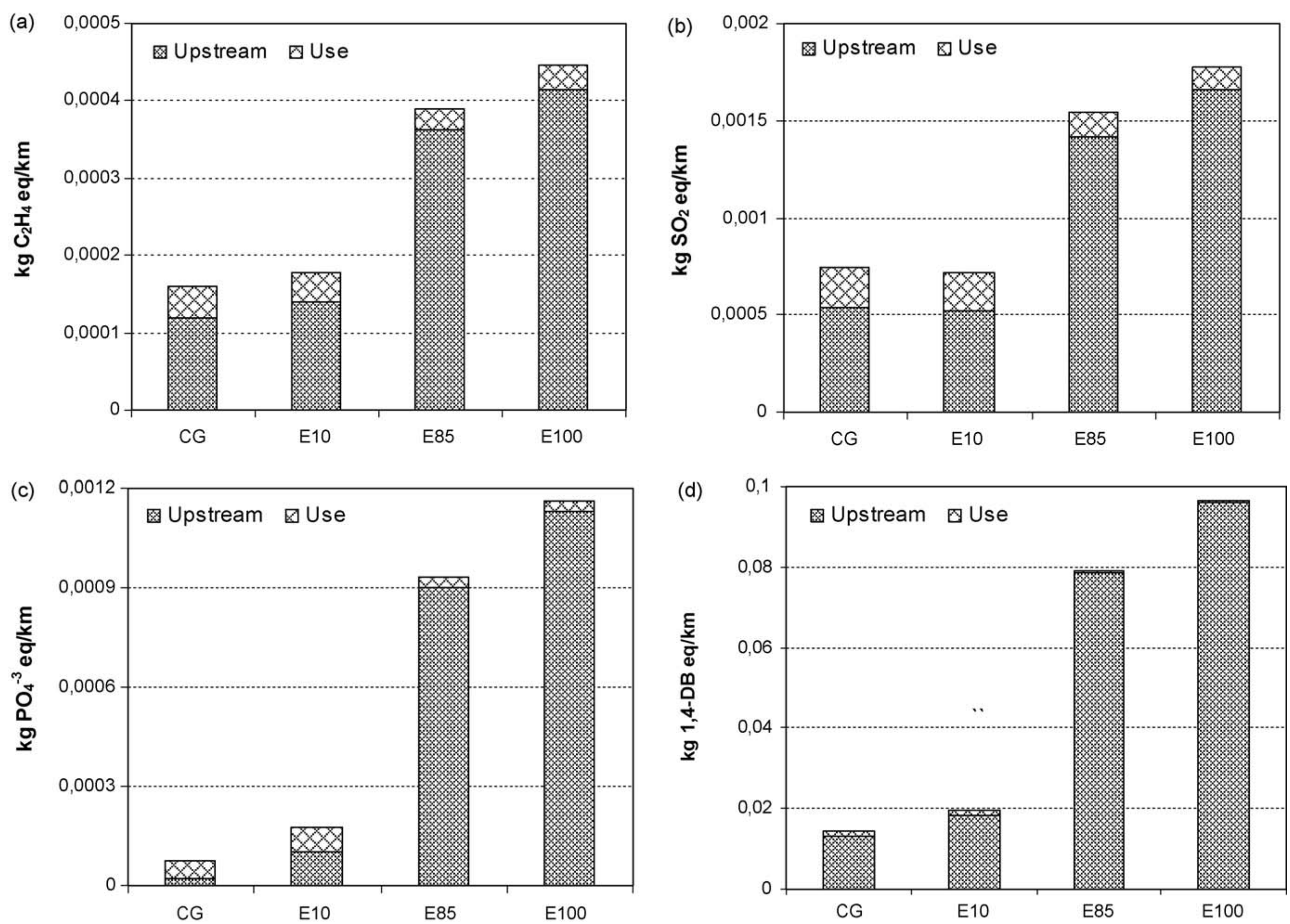

Fig. 5. Net life cycle air emissions for CG, E10, E85 and E100 based on mass allocation. (a) Contributions to photochemical oxidants formation; (b) contributions to acidification; (c) contributions to eutrophication; (d) contributions to human toxicity.

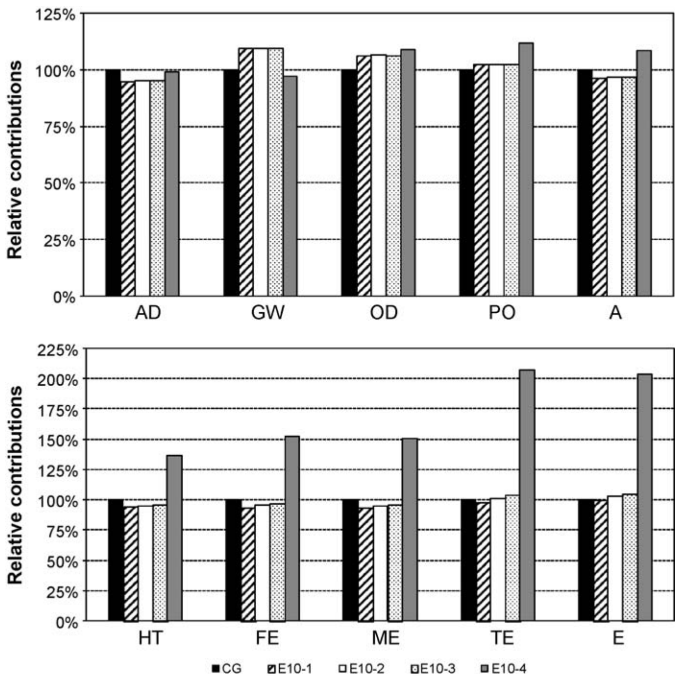

Fig. 6. Relative environmental profile of the compared E10 scenarios, with CG being the baseline (index = 100). Allocation acronyms: (1) economic allocation 1; (2) economic allocation 2; (3) economic allocation 3; (4) mass allocation.

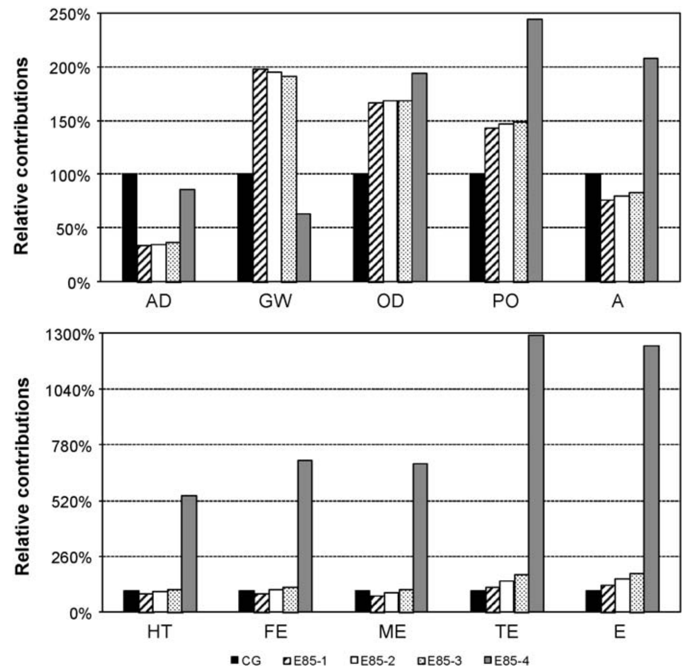

Fig. 7. Relative environmental profile of the compared E85 scenarios, with CG being the baseline (index = 100). Allocation acronyms: (1) economic allocation 1; (2) economic allocation 2; (3) economic allocation 3; (4) mass allocation.

comprise only co-products such as biogas, syrup and solid wastes) in all the life cycle. Despite the increase of CH_4 and N_2O emissions, decrease of CO_2 emissions make ethanol based fuels more climate friendly than CG. Numerous authors have previously reported the benefits of using ethanol based fuels in order to reduce GHG emissions, subsequently reducing the effects on climate change and pollution [48]. Therefore, the contributions to GW were analysed in more detail for MA since it is the only scenario where a reduction of GHG emissions is achieved. In addition, the main processes responsible for the environmental performance of each fuel were identified and are shown in Fig. 4. According to our results, gasoline production and use is the main hot spot in terms of GW for CG. Regarding E10, ethanol based fuel use is the main hot spot followed by gasoline production (90% in volume of the blend). The ethanol conversion stage (mainly the electricity production process) presents a small contribution. Nevertheless, diesel production is increased (included in 'others') because of the increase in agricultural activities. Electricity production and distillation, and ethanol use in FFV were identified as hot spots for E85 and E100 blends, due to CO_2 emissions. In addition, it is necessary to comment the contribution of diesel production consumed in field activities due to the high rate of ethanol employed in both fuels.

Environmental burdens from upstream activities are the main responsible in almost all impact categories and have higher contributions in ethanol based blends than when CG is used. This is due to feedstock related activities such as the fertilizers production and use, combustion emissions from agricultural machinery and machinery production. Fig. 5 confirms these results in terms of PO, A, E and HT. FE, ME and TE were not included in the figure since almost 100% of total contributions are due to upstream activities (mainly production of agricultural machinery and oil extraction in ethanol blends and gasoline, respectively).

3.2.2. Comparison between allocation procedures

The purpose of the sensitivity analysis is to estimate the effects on the outcomes of a study of the allocation procedures. Figs. 6–8 present the life cycle environmental performance of E10 fuels, E85 fuels and E100 fuels according to the sensitivity analysis for the change of the allocation ratio between shives and the other agricultural products based on economic and mass approaches, in comparison with CG. All corresponding results are displayed as percentages relative to CG.

As seen in all the figures, changes in the results represent the effect of changing the allocation factors. According to Fig. 6,

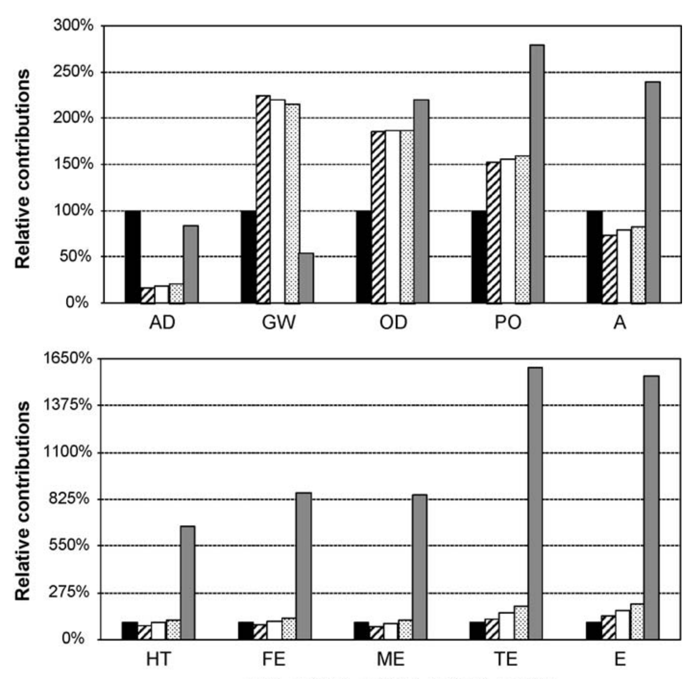

Fig. 8. Relative environmental profile of the compared E100 scenarios, with CG being the baseline (index = 100). Allocation acronyms: (1) economic allocation 1; (2) economic allocation 2; (3) economic allocation 3; (4) mass allocation.

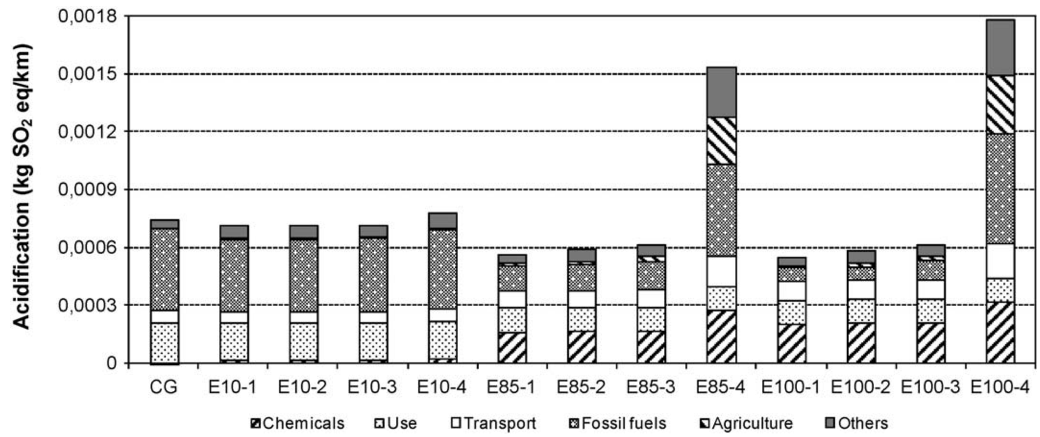

Fig. 9. Comparative life cycle SO_2 equivalent emissions and main processes involved for CG, E10, E85 and E100 per functional unit. Acronyms: (1) EA1; (2) EA2; (3) EA3; (4) MA.

shifting from CG to E10 fuels only presents environmental benefits in terms of AD regardless of the allocation factor chosen. The higher the reduction of the contributions to AD is, the lower the allocation factor is. In this impact category, fossil fuel extraction is the main hot spot and it is reduced due to (i) less diesel from agricultural activities is allocated to ethanol when the allocation factor is reduced and (ii) there is less gasoline in the transport fuel than when CG is used. In addition, GHG emissions could be reduced using E10 as liquid fuel. However, this is only possible when the allocation factor is high; that is, when mass allocation is considered. The reason for this is the fact that more C sequestration is assigned to the flax shives, obtaining offset in GHG emissions for the remaining processes. According to our results, contributions to toxicity categories such as HT, FE, ME and A are also reduced with regard to the use of CG only in economic allocation caused by the low contribution of agricultural activities to ethanol production (almost negligible). The result is completely opposite for mass allocation and CG shows a better environmental profile. With regard to the remaining impact categories (OD, PO, E and TE), results from economic allocations are better than from mass allocation although in all cases, the environmental profile is worse than when CG is used as transport fuel. The adverse effects are mainly caused because of environmental burdens released during flax cultivation (mainly, nitrogen and phosphorous related emissions), combustion emissions from agricultural machinery and fertilizer production.

According to Figs. 7 and 8, once again economic allocations offer better environmental performance than mass allocation in all impact categories excluding GW. Reduction of up to almost 50% can be achieved in terms of GHG emissions when mass allocation is taken into account as well as E100 fuel is used as transport fuel.

With regard to A, HT and some ecotoxicity categories the advantages of using ethanol blends can be highlighted (independently of the ratio of ethanol in the blend), but only when low allocation factors are assumed for the feedstock (economic allocation). These credits are more remarkable in HT and A, and therefore, they have been analysed in further detail. Figs. 9 and 10 show the comparison between scenarios and the main stages involved in both impact categories: chemical production, end use, transport systems involved in all the life cycle, fossil fuel extraction, agricultural activities and others. Others include activities such as infrastructure construction, agricultural machinery production and waste treatment. According to Fig. 9, fossil fuel extraction is the main hot spot in CG and E10 fuels, followed by their use. There is almost no contribution from agricultural activities and chemical production in E10 fuels, especially for economic allocation. SO_2 equivalent emissions from the use are independent of the allocation factor but, emissions from the other stages show a high dependency on the allocation factors. It can be seen in E85 and E100 that there are large differences in the SO_2 equivalent emissions according to the allocation factors used. In both fuels, the contributions from chemical production and agriculture are higher. The magnitude of the increase is proportional to the allocation factor for the shives. The effect is very similar when HT is analysed. According to Fig. 10, fossil fuel extraction is the hot spot in CG and E10 but, activities related to

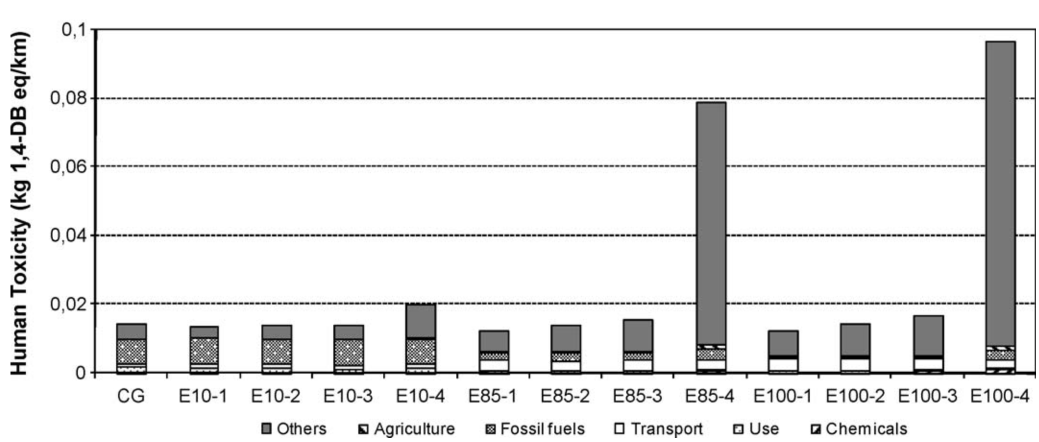

Fig. 10. Comparative life cycle 1,4-DB equivalent emissions and main processes involved for CG, E10, E85 and E100 per functional unit. Acronyms: (1) EA1; (2) EA2; (3) EA3; (4) MA.

infrastructure are the main hot spots in E85 and E100 fuels. These contributions get larger when the allocation factor is increased. Regarding the ecotoxicity impact categories, infrastructure related activities are the hot spots in all fuels regardless to the allocation factor. The contributions from feedstock cultivation are really low (1–9%) for high allocation factors (mass allocation).

4. Discussion

The substitution of fossil fuels with biofuels has been proposed by the EU as part of a strategy to mitigate GHG emissions from road transport, increase security in the energy supply and support development of rural communities. Several environmental studies related to second generation ethanol production and use suggest that this kind of fuels could have the potential to mitigate climate change and reduce dependency on fossil fuels [48,49]. However, sensitivity analyses show the influence in the results of factors such as the choice of the functional unit [7], the use or not of biomass wastes as fuel in the ethanol plant [3,26,29] or the choice of the allocation factors [50].

Choosing the allocation factor is a key point in the lignocellulosic ethanol production cycle since lignocellulosic feedstocks usually are inexpensive and abundant waste products from the forestry industry and agricultural residues. Therefore, the selection of the allocation procedure (mass, economic, energy...) could alter the final environmental results.

This paper presented an explanation of the allocation choice taking into account two allocation procedures. On the one hand, there are large differences in the economic value of the agricultural products obtained from flax crops. Flax shives present a relatively low market price (around 0.026 €/kg compared to 2.18 €/kg for long fibres). On the other hand, large amounts of shives are generated (roughly 67.5% of total biomass produced) in flax crops although the motivating force is the production of fibres (~25%). Therefore, allocations based on weight and several market values were selected.

The use of ethanol based fuels present advantages and disadvantages, such as transport fuel in a FFV. This paper focuses on identifying them, taking into account the effect from the allocation approach chosen.

When mass allocation was applied, impact indicators like GW and AD were lower for the ethanol containing fuels (per km driven), whereas the remaining impact indicators showed higher values. According to our results, equivalent CO_2 emission savings of 7, 95 and 118 g/km were achieved for E10, E85 and E100, respectively. These results fit in with previously published studies. Fu et al. [3] reported that E10 could reduce in 8 g/km the equivalent CO_2 emission when waste biomass is used as feedstock and energy source in the plant. Kim and Dale [7] reported savings of 15 and 140 g/km in E10 and E85 respectively when corn is used as the raw material and Ryan et al. [51] accounted savings of up to 191 g/km when lignocellulosic residues based ethanol is used. Luo et al. [27] reported a reduction of 62% in equivalent CO_2 emissions per km driven. However, this final computation is a function of the sort of raw material cultivated, the agricultural yield, the use of products and co-products and the economic value. Regarding fossil fuels depletion, our results are similar to those obtained by other authors [3,7,26,29]. Reductions of almost 45% in fossil fuel extraction can be achieved when E100 from flax shives is used as fuel, considering mass allocation. Luo et al. [27] achieved reductions of 50% when corn stover was used as feedstock and mass allocation.

However, the results are completely different when economic allocation is considered as the base of our study. This explains the importance of the selection of the allocation factors. GW should be increased using ethanol as liquid fuel. However, in the remaining impact categories the environmental performance of the ethanol based fuels use is considerably better than when mass allocation is used. These opposite effects are mainly due to the burdens related to flax cultivation, particularly the lowest contribution of agricultural activities. In fact, contributions to acidification (one of the most important categories affected by biomass based fuels) should be reduced up to 25% with regard to the use of conventional gasoline. Furthermore, reductions of up to 75% in fossil fuel extraction can be achieved when both E100 and economic allocation are taken into account due to the absence of gasoline in the transport fuel and the negligible presence of feedstock cultivation step.

Even though this study is relatively thorough, there are some limitations to the work due to the fact that flax shives are not used nowadays as commercial ethanol production feedstock and cellulosic ethanol conversion technology is under development.

5. Conclusions

This study shows the results of a LCA performed upon flax shives based fuel ethanol and its use in FFV whether blended or not with gasoline. In this study, flax shives are agricultural co-products from pulp fibre production (the main product of this kind of crop). Shive production, processing and transport to refinery gate, ethanol conversion and transport to blending refinery, ethanol blending with gasoline in two ethanol fuel applications (E10 and E85) and, E10, E85 and E100 burning in FFV were evaluated and compared with the use of conventional gasoline.

According to the results, the allocation methodology can greatly alter the environmental effects when different alternatives are being compared. This illustrates the importance of avoiding allocation when possible, as the selection of a coefficient potentially affects the conclusions of the study.

Cellulosic fuel ethanol as a blend with gasoline (or not) may help to reduce the greenhouse gases emissions only when a large allocation factor is assumed for the flax shives (in this case, mass allocation). However, using ethanol based fuels would increase the contributions to other impact categories, such as eutrophication and photochemical oxidants formation and should reduce the fossil fuel consumption in spite of the allocation factor selected. On the contrary, the contributions to other impact categories such as acidification, human toxicity and ecotoxicity could be reduced when a small allocation factor is assumed due to the shorter contributions from the feedstock cultivation stage.

Ethanol fuel used in form of blends in gasoline can help increase the security in the energy supply regardless the allocation coefficient chosen. Agricultural activities related to feedstock production were identified as notable contributors to the environmental performance. Thus, high yielding varieties, reduction of tillage activities and decline in fertilization should help to reduce these impacts.

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